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The drivers and health risks of unexpected surface ozone enhancements over the Sichuan Basin, China, in 2020

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Section S1. Data filter criteria

We applied a data quality control method used in Lu et al. (2020) to remove unreliable ozone data. Hourly observed data points were transformed into Z scores, and then, the observed data were removed if the corresponding Z_i met one of the following conditions: (1) Z_i is larger or smaller than the previous one (Z_{i-1}) by 9 ($|Z_i - Z_{i-1}| > 9$), (2) The absolute value of Z_i is greater than 4 ($|Z_i| > 4$), or (3) the ratio of the Z value to the third-order center moving average is greater than 2 ($\frac{3Z_i}{Z_{i-1}+Z_i+Z_{i+1}} > 2$). The formula for calculating Z_i is as follows:

$$Z_i = \frac{X_i - \bar{X}}{\sigma} \tag{1}$$

where X_i represents the i-th item in the dataset, and $\bar{X}$ and σ are the average and standard deviation of dataset X, respectively. The distribution of CNMEC sites over the SCB is shown in Figure 1.

Section S2. Metrics definitions

As the parameters listed in Table S1 are different in units and magnitudes, which could lead to unstable performance of the training model. Therefore, we standardized all the parameters before using them for model training. The standardized process is expressed as below:

$$D_i = \frac{P_i - \mu}{\sigma} \tag{2}$$

where P_i, μ, and σ are the i-th parameter, the average, and the standard deviation of the training input dataset listed in Table S1, respectively. D_i represents the standardized value used for model training.

The root-mean-square error (RMSE), normalized mean bias (NMB), normalized root-mean-square error (NRMSE), and Pearson correlation coefficient (R) are used to evaluate the performance of the GEOS-Chem-XGBoost model. The formulas of these metrics are as follows:

$$RMSE = \sqrt{\frac{1}{N}\sum_{i=0}^{n}(X_i - Y_i)^2} \tag{3}$$

$$NMB = \frac{\sum_{i=0}^{n}(X_i - Y_i)}{\sum_{i=0}^{n} Y_i} \tag{4}$$

$$NRMSE = \frac{\sqrt{\frac{1}{N}\sum_{i=0}^{n}(X_i - Y_i)^2}}{Y_{95th} - Y_{5th}} \tag{5}$$

$$R = \frac{N\sum_{i=0}^{n} X_i Y_i - \sum_{i=0}^{n} X_i \cdot \sum_{i=0}^{n} Y_i}{\sqrt{N\sum_{i=0}^{n} X_i^2 - \left(\sum_{i=0}^{n} X_i\right)^2} \cdot \sqrt{N\sum_{i=0}^{n} Y_i^2 - \left(\sum_{i=0}^{n} Y_i\right)^2}} \tag{6}$$

where X and Y represent the datasets of model and measurement, respectively.

Table S1. List of input parameters fed into each XGBoost machine learning model.

Acronym	Description
Concentrations of chemical constituents simulated by GEOS-Chem model	
NO_2_geos	Nitrogen dioxide
NO_geos	Nitrogen oxide
NO_x_geos	Reactive nitrogen (NO+NO_2+nitrates)
O_3_geos	Ozone
CO_geos	Carbon monoxide
ACET_geos	Acetone
ALK4_geos	Alkanes
ALD2_geos	Acetaldehyde
HCHO_geos	Formaldehyde
C_2H_6_geos	Ethane
C_3H_8_geos	Propane
BCPI_geos	Hydrophilic black carbon
BCPO_geos	Hydrophobic black carbon
OCPI_geos	Hydrophilic organic carbon
OCPO_geos	Hydrophobic organic carbon
EOH_geos	Ethanol
DST1_geos	Dust with diameter of 0.7μm
DST2_geos	Dust with diameter of 1.4μm
DST3_geos	Dust with diameter of 2.4μm
DST4_geos	Dust with diameter of 4.5μm
H_2O_2_geos	Hydrogen peroxide
HNO_3_geos	Nitric acid
HNO_4_geos	Pyroxenitic acid
ISOP_geos	Isoprene
MACR_geos	Methacrolein
MEK_geos	Methyl ethyl ketone
MVK_geos	Methyl vinyl ketone
N_2O_5_geos	Dinitrogen pentoxide
NH_3_geos	Ammonia
NH_4_geos	Ammonium
NIT_geos	Inorganic nitrates
PAN_geos	Peroxyacetyl nitrate
PRPE_geos	Alkenes
RCHO_geos	Aldehyde
SALA_geos	Fine sea salt aerosol
SALC_geos	Coarse sea salt aerosol
SO_2_geos	Sulfur dioxide
SOAP_geos	Secondary organic aerosol precursor
SOAS_geos	Simple secondary organic aerosol
TOLU_geos	Toluene
XYLE_geos	Xylene

O_x_geos	Odd oxygen (O_3 + NO_2)
$PM_{2.5}$_geos	$PM_{2.5}$
Emissions used for GEOS-Chem simulation	
ENO	Nitrogen oxide emissions
ECO	Carbon monoxide emissions
EACET	Acetone emissions
EALD2	Acetaldehyde emissions
EALK4	Alkanes emissions
EBENZ	Benzene emissions
EC_2H_6	Ethane emissions
EPRPE	Alkenes emissions
ETOLU	Toluene emissions
EXYLE	Xylene emissions
EISOP	Isoprene emissions
EBCPI	Hydrophilic black carbon emissions
EBCPO	Hydrophobic black carbon emissions
EOCPI	Hydrophilic organic carbon emissions
EOCPO	Hydrophobic organic carbon emissions
ESALA	Fine sea salt aerosol emissions
ESALC	Coarse sea salt aerosol emissions
ESO_2	Sulfur dioxide emissions
ESOAP	Secondary organic aerosol precursor emissions
ESOAS	Simple secondary organic aerosol emissions
$ECHBR_3$	Bromoform emissions
Time information	
Hour	Hour of day
Weekday	Day of the week
Month	Month of the year
Trendday	Days since 1 May 2019
Meteorological fields used for GEOS-Chem simulation	
Cldtt	Total cloud fraction
Ps	Surface pressure
Q_{10m}	Specific humidity at 10m
Q_{2m}	Specific humidity at 2m
T_{10m}	Temperature at 10m
Tprec	Total precipitation
T	Skin surface temperature
T_{2m}	Temperature at 2m
U_{10m}	10m East-West wind-speed
V_{10m}	10m North-South wind-speed
U	Skin surface East-West wind-speed
V	Skin surface North-South wind-speed
Zpbl	Planetary boundary layer height

Table S2. Daily y_0 and β values for all non-accidental causes, cardiovascular diseases (CVD), respiratory diseases (RD), hypertension, stroke and chronic obstructive pulmonary disease (COPD). This table is cited from Wang et al. (2021).

Disease	Daily y_0	β (%)
All non-accidental causes	1.687×10^{-5}	0.24
CVD	3.880×10^{-6}	0.27
RD	1.841×10^{-6}	0.18
Hypertension	5.422×10^{-7}	0.60
Stroke	1.197×10^{-7}	0.29
COPD	1.623×10^{-6}	0.20

Table S3. Anthropogenic emissions of NO_x and VOCs for Chongqing and Sichuan provinces in May-June 2019 and 2020 provided by the MEE over the SCB (Unit: Tonne)

Pollutant	Month	Sector	2019		2020		% Difference	
			Chongqing	Sichuan	Chongqing	Sichuan	Chongqing	Sichuan
NO_x	May	power	3971	3130	6042	5048	52.15%	61.28%
		industry	11681	30494	11759	29505	0.67%	-3.24%
		residential	956	2443	970	2485	1.46%	1.72%
		transportation	13138	21253	12290	20017	-6.45%	-5.82%
		agriculture	0	0	0	0	0%	0%
		total	29746	57320	31061	57055	4.50%	-0.46%
	June	power	3732	3446	4814	3791	28.99%	10.01%
		industry	13384	35479	13623	34402	1.79%	-3.04%
		residential	924	2362	937	2402	1.41%	1.69%
		transportation	13138	21253	12300	19809	-6.38%	-6.79%
		agriculture	0	0	0	0	0%	0%
		total	31178	62540	31674	60404	-1.59%	-3.41%
VOCs	May	power	34	25	52	40	52.94%	60.00%
		industry	27938	67606	27029	63614	-3.25%	-5.90%
		residential	6229	16419	6100	16150	-2.07%	-1.64%
		transportation	5732	15861	5389	14348	-5.98%	-9.54%
		agriculture	0	0	0	0	0%	0%
		total	39933	99911	38570	94152	-3.41%	-5.67%
	June	power	32	27	41	30	28.13%	11.11%
		industry	34528	83393	33523	78503	-2.91%	-5.86%
		residential	6069	15967	5946	15708	-2.03%	-1.62%
		transportation	5732	15861	6016	16211	4.95%	2.21%
		agriculture	0	0	0	0	0%	0%
		total	46361	115248	45526	110452	-1.80%	-4.16%

Table S4. Total daily mortalities from all non-accidental causes (NAC), CVD, RD, COPD, hypertension (HT), and stroke attributable to ambient O_3 exposure in each city over the SCB within May-June and the whole year in 2019.

City	Period	NAC	CVD	RD	HT	Stroke	COPD
Abazhou	Jan-Dec	86	22	7	7	1	7
	May-Jun	8	2	1	1	0	1
Bazhong	Jan-Dec	271	70	22	21	2	22
	May-Jun	49	13	4	4	0	4
Chengdu	Jan-Dec	3427	882	283	259	29	277
	May-Jun	850	219	70	66	7	68
Chongqing	Jan-Dec	4851	1248	402	365	41	392
	May-Jun	415	107	34	32	4	33
Dazhou	Jan-Dec	564	145	46	43	5	45
	May-Jun	31	8	3	2	0	2
Deyang	Jan-Dec	529	136	44	40	4	43
	May-Jun	124	32	10	10	1	10
Ganzizhou	Jan-Dec	31	8	3	2	0	2
	May-Jun	0	0	0	0	0	0
Guangan	Jan-Dec	415	107	34	32	4	33
	May-Jun	80	21	7	6	1	6
Guangyuan	Jan-Dec	141	36	12	11	1	11
	May-Jun	10	3	1	1	0	1
Leshan	Jan-Dec	442	114	36	34	4	36
	May-Jun	87	22	7	7	1	7
Liangshanzhou	Jan-Dec	934	241	77	71	8	75
	May-Jun	246	63	20	19	2	20
Luzhou	Jan-Dec	559	144	46	42	5	45
	May-Jun	40	10	3	3	0	3
Meishan	Jan-Dec	571	147	47	43	5	46
	May-Jun	142	36	12	11	1	11
Mianyang	Jan-Dec	664	171	55	51	6	54
	May-Jun	164	42	13	13	1	13
Nanchong	Jan-Dec	656	169	54	50	6	53
	May-Jun	134	35	11	11	1	11
Neijiang	Jan-Dec	407	105	34	31	3	33
	May-Jun	60	15	5	5	1	5
Panzhihua	Jan-Dec	208	54	17	16	2	17
	May-Jun	62	16	5	5	1	5
Suining	Jan-Dec	353	91	29	27	3	28
	May-Jun	60	16	5	5	1	5
Yaan	Jan-Dec	170	44	14	13	1	14
	May-Jun	33	8	3	3	0	3
Yibin	Jan-Dec	734	189	61	56	6	59

	May-Jun	123	32	10	10	1	10
Zigong	Jan-Dec	391	101	32	30	3	32
	May-Jun	71	18	6	6	1	6
Ziyang	Jan-Dec	371	96	31	28	3	30
	May-Jun	87	22	7	7	1	7
Total	Jan-Dec	16773	4319	1386	1272	143	1354
	May-Jun	2874	741	237	223	25	231

Table S5. Total daily mortalities from all non-accidental causes (NAC), CVD, RD, COPD, hypertension (HT), and stroke attributable to ambient O_3 exposure in each city over the SCB within May-June and the whole year in 2020.

City	Peoriod	NAC	CVD	RD	HT	Stroke	COPD
Abazhou	Jan-Dec	75	19	6	6	1	6
	May-Jun	11	3	1	1	0	1
Bazhong	Jan-Dec	273	70	22	21	2	22
	May-Jun	95	25	8	7	1	8
Chengdu	Jan-Dec	4376	1126	362	328	37	354
	May-Jun	1488	383	123	113	13	120
Chongqing	Jan-Dec	4374	1126	362	331	37	353
	May-Jun	1070	276	88	83	9	86
Dazhou	Jan-Dec	411	106	34	32	4	33
	May-Jun	57	15	5	4	0	5
Deyang	Jan-Dec	657	169	54	50	6	53
	May-Jun	220	57	18	17	2	18
Ganzizhou	Jan-Dec	59	15	5	5	1	5
	May-Jun	10	2	1	1	0	1
Guangan	Jan-Dec	522	135	43	40	4	42
	May-Jun	156	40	13	12	1	13
Guangyuan	Jan-Dec	287	74	24	22	2	23
	May-Jun	110	28	9	8	1	9
Leshan	Jan-Dec	546	141	45	41	5	44
	May-Jun	181	47	15	14	2	15
Liangshanzhou	Jan-Dec	814	210	67	63	7	66
	May-Jun	183	47	15	14	2	15
Luzhou	Jan-Dec	674	174	56	51	6	54
	May-Jun	164	42	13	13	1	13
Meishan	Jan-Dec	639	164	53	48	5	52
	May-Jun	205	53	17	16	2	17
Mianyang	Jan-Dec	868	224	72	66	7	70
	May-Jun	304	78	25	23	3	24
Nanchong	Jan-Dec	595	154	49	46	5	48
	May-Jun	174	45	14	14	1	14
Neijiang	Jan-Dec	547	141	45	42	5	44

	May-Jun	172	44	14	13	1	14
Panzhihua	Jan-Dec	178	46	15	14	2	14
	May-Jun	47	12	4	4	0	4
Suining	Jan-Dec	422	109	35	32	4	34
	May-Jun	132	34	11	10	1	11
Yaan	Jan-Dec	174	45	14	13	1	14
	May-Jun	63	16	5	5	1	5
Yibin	Jan-Dec	839	216	69	64	7	68
	May-Jun	269	69	22	20	2	22
Zigong	Jan-Dec	503	129	42	38	4	41
	May-Jun	169	44	14	13	1	14
Ziyang	Jan-Dec	465	120	38	35	4	38
	May-Jun	177	46	15	13	2	14
Total	Jan-Dec	18301	4712	1513	1387	156	1477
	May-Jun	5455	1406	450	418	46	440

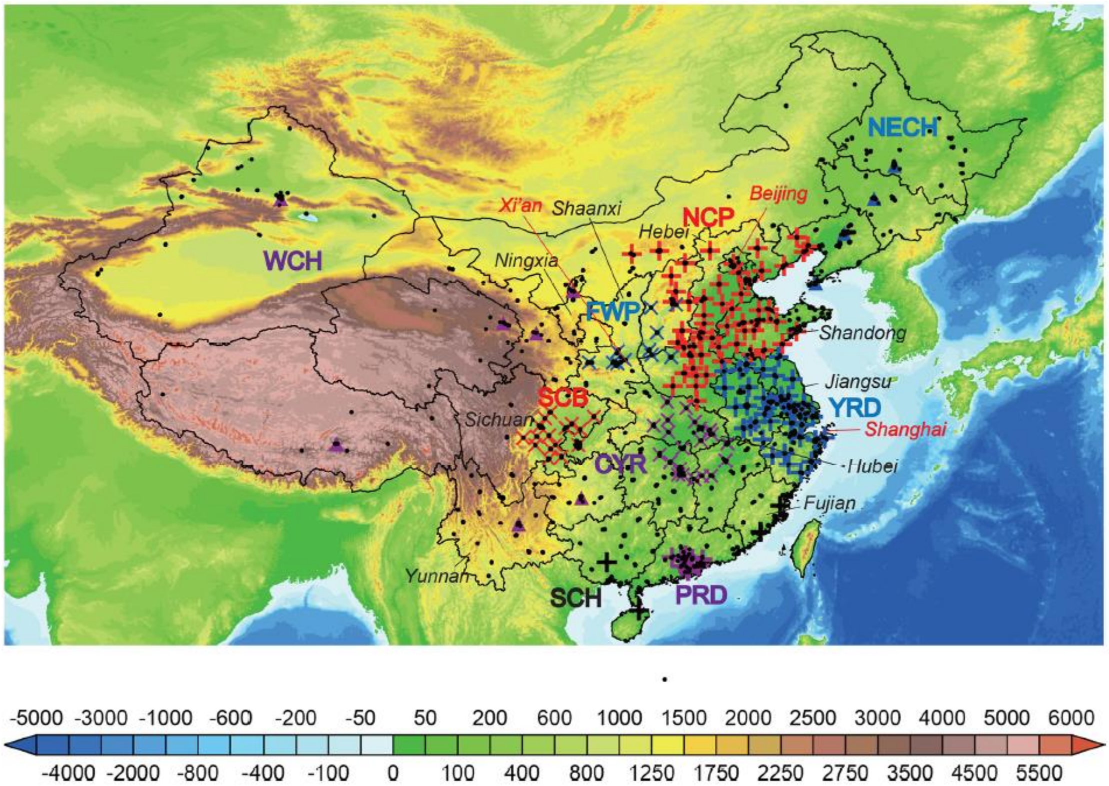

Figure S1. Site locations (black dots) of the China National Environmental Monitoring Center (CNEMC) network. Also shown are the locations of 169 major cities grouped to 9 city clusters: the North China Plain (NCP; red pluses) cluster, the Yangtze River Delta (YRD; blue pluses) cluster, the Fenwei Plain (FWP; blue crosses) cluster, the Sichuan Basin (SCB; red crosses) cluster, the central Yangtze River (CYR; purple crosses) cluster, the Pearl River Delta (PRD; purple pluses) cluster, the northeastern China (NECH; blue triangles) cluster, the western China (WCH; purple triangles) cluster, and southern China (SCH; black pluses). The underlying figure shows terrain elevation (m). This figure is cited from Lu et al. (2019).

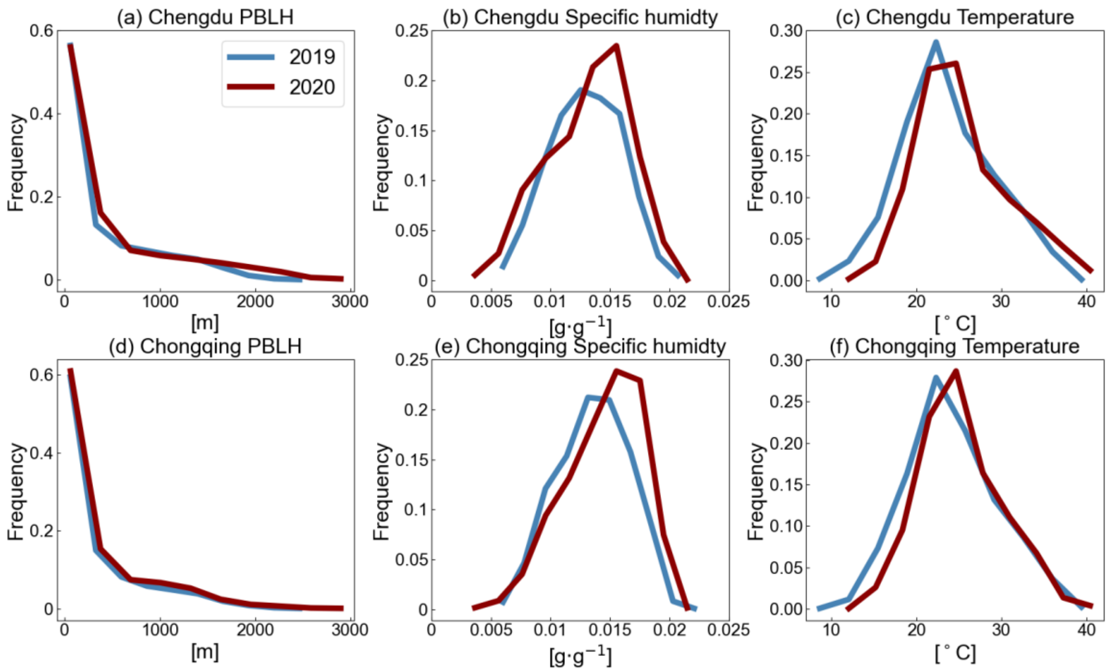

Figure S2. Probability density functions (PDFs) of hourly planetary boundary layer height (PBLH), temperature at 2 m, and relative humidity in the whole 2019 (blue) and May-June 2020 (red) at Chengdu and Chongqing cities over the SCB, from the GEOS-FP meteorology fields that are used to drive the GEOS-Chem model and to train/predict the ozone bias. We group each data to 10 bins, and frequencies are calculated for each bin.

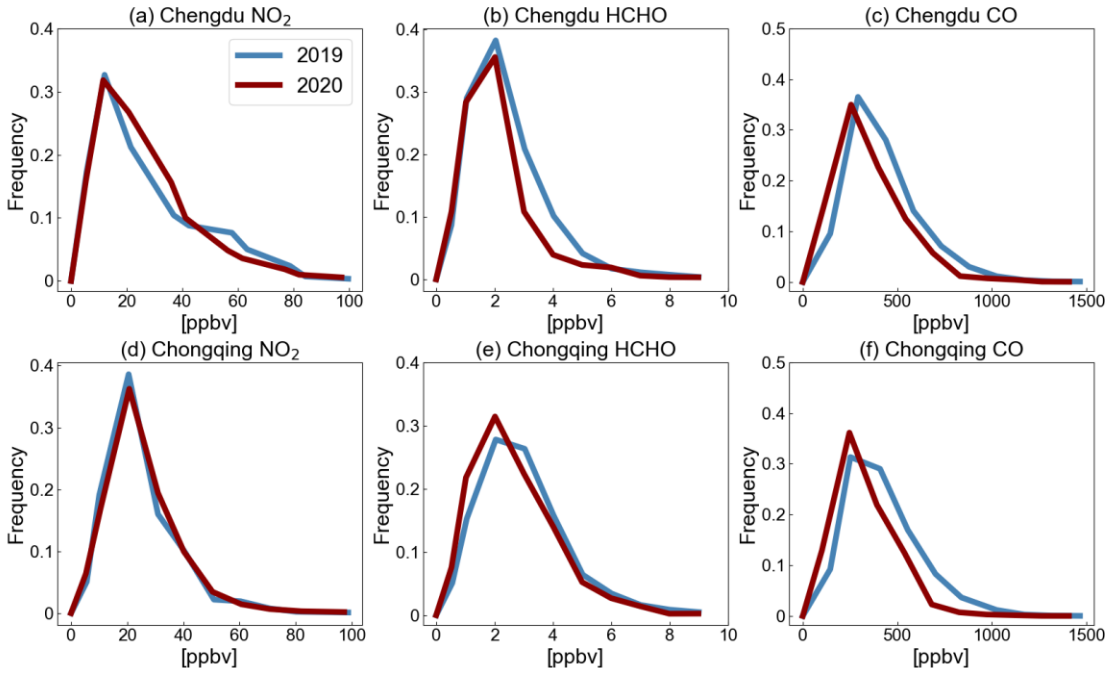

Figure S3. Same as Figure S2, but for hourly concentrations of GEOS-Chem NO_2, CO, and HCHO. The difference between 2019 and 2020 in GEOS-Chem only reflects meteorological effects.

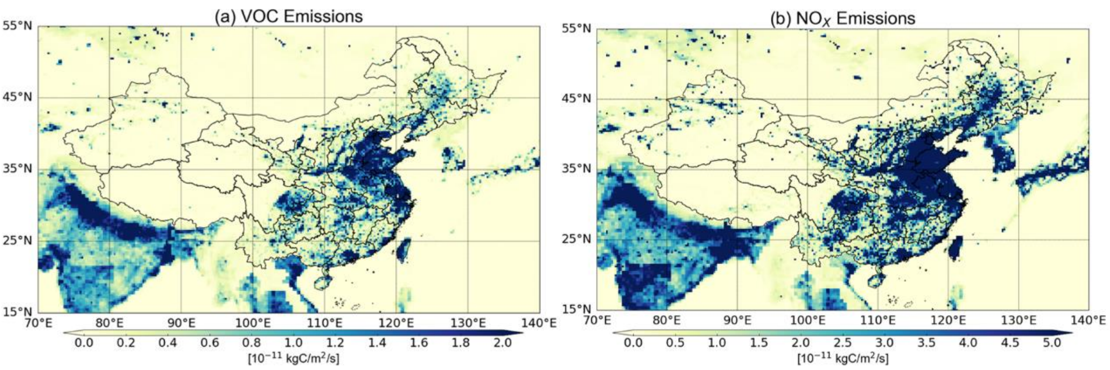

Figure S4. The distribution of anthropogenic emissions of VOCs and NO_x across China prescribed from the MEIC inventory v2017. The base map of the figure is created by the Basemap package of Python.

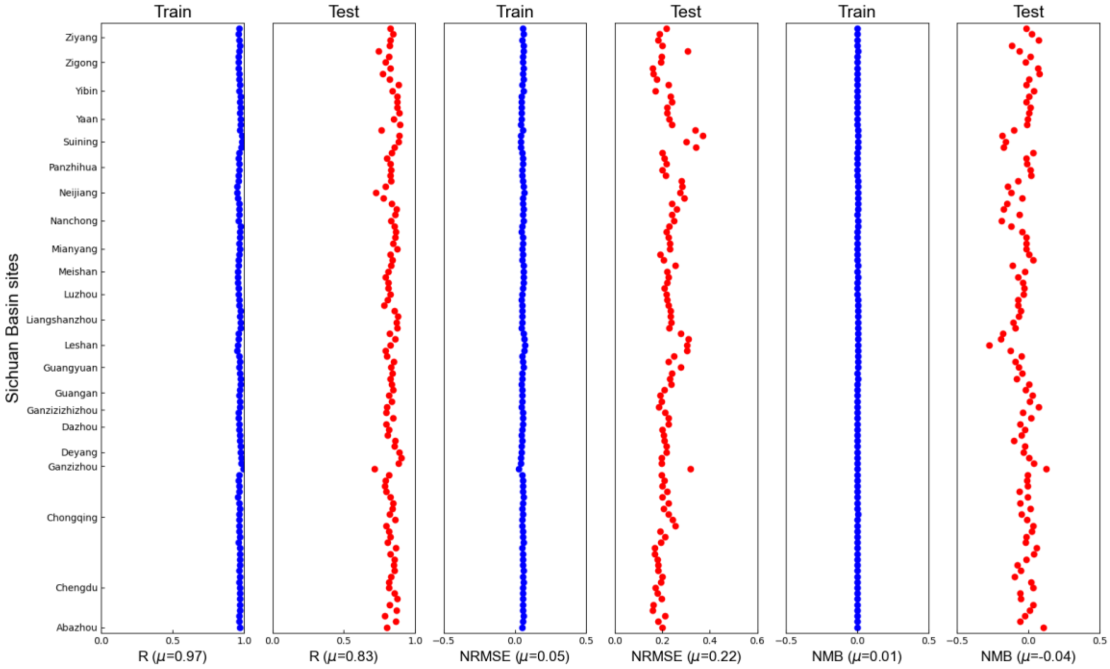

Figure S5. Statistics for the performance of GEOS-Chem-XGBoost model to predict hourly O_3 measurements over the SCB in 2019. Shown are Pearson correlation coefficients (R), normalized root-mean-square error (NRMSE), and the normalized mean bias (NMB), for the training data (blue) and the test data (red).

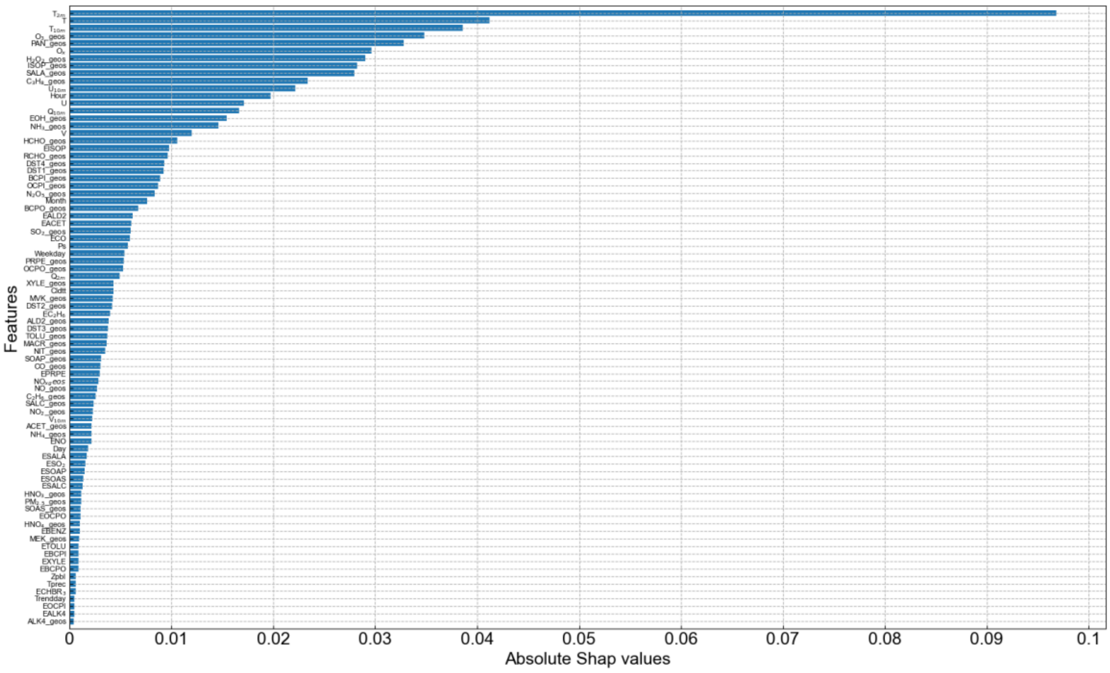

Figure S6. The same as Figure 2 but for all variables.

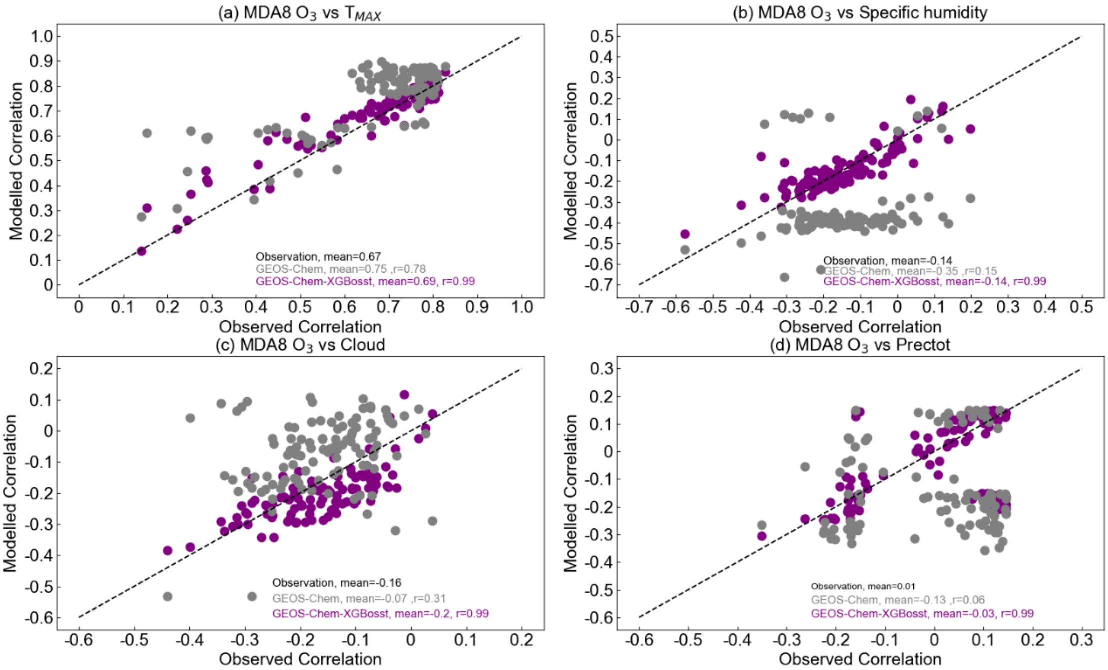

Figure S7. Measured and modelled correlation coefficients between deseasonalized surface MDA8 ozone and daily maximum 2 m temperature $r_{ozone,T}$ (a), specific humidity $r_{ozone,H}$ (b), cloud fraction $r_{ozone,C}$ (c), and precipitation $r_{ozone,P}$ (d). The mean $r_{ozone,T}$, $r_{ozone,H}$, $r_{ozone,C}$, and $r_{ozone,P}$ values, and the correlation coefficients between observations and GEOS-Chem and between observations and GEOS-Chem-XGBoost results are shown inset.

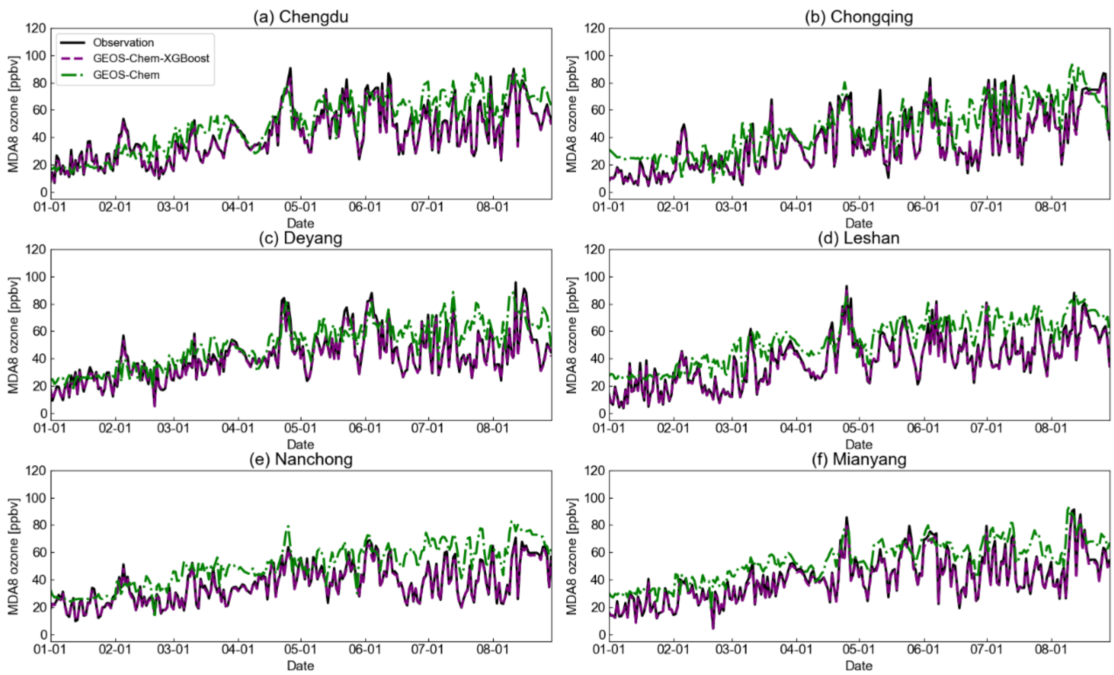

Figure S8. Measured and modelled O_3 variabilities in the selected cities over the SCB in 2019. Measured, GEOS-Chem, and GEOS-Chem-XGBoost predicted O_3 concentrations are denoted by black solid, green dashed, and purple dashed lines, respectively. (a) Chengdu, (b) Chongqing, (c) Deyang, (d) Leshan, (e) Nanchong, and (e) Mianyang.

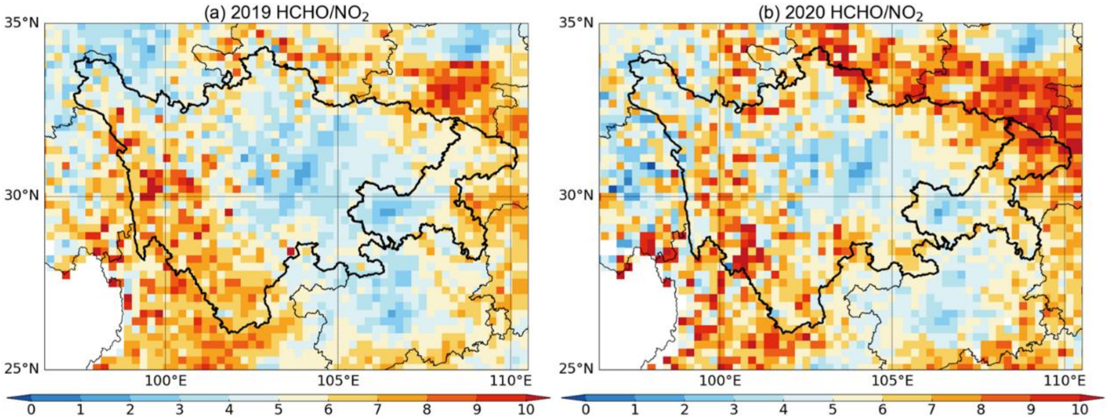

Figure S9. May-June mean TROPOMI $HCHO/NO_2$ ratios between (a) 2019 and (b) 2020. The $HCHO/NO_2$ ratios in May-June in most cities over the SCB have indicated a shift toward high values from 2019 to 2020 but the O_3 chemical sensitivities in 2020 in most cities still lie within the transitional regime between 1 and 7. The base map of the figure is created by the Basemap package of Python.

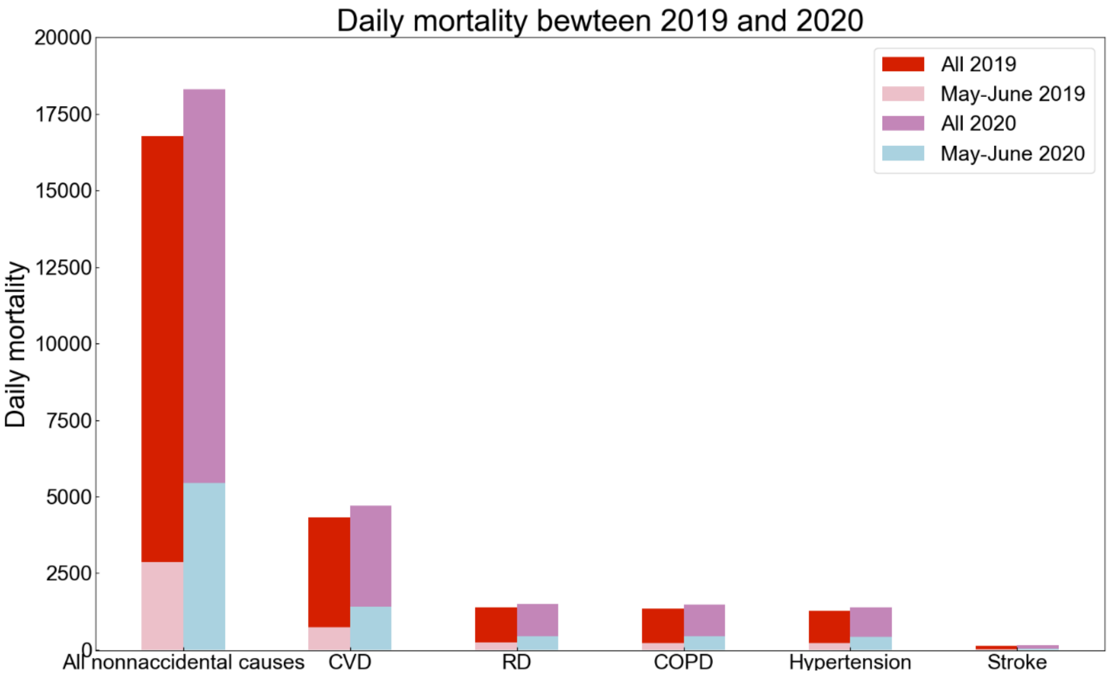

Figure S10. Total daily mortality from all non-accidental causes, CVD, RD, COPD, hypertension, and stroke attributable to ambient O_3 exposure in all cities over the SCB within May-June and the whole year in 2019 and 2020.

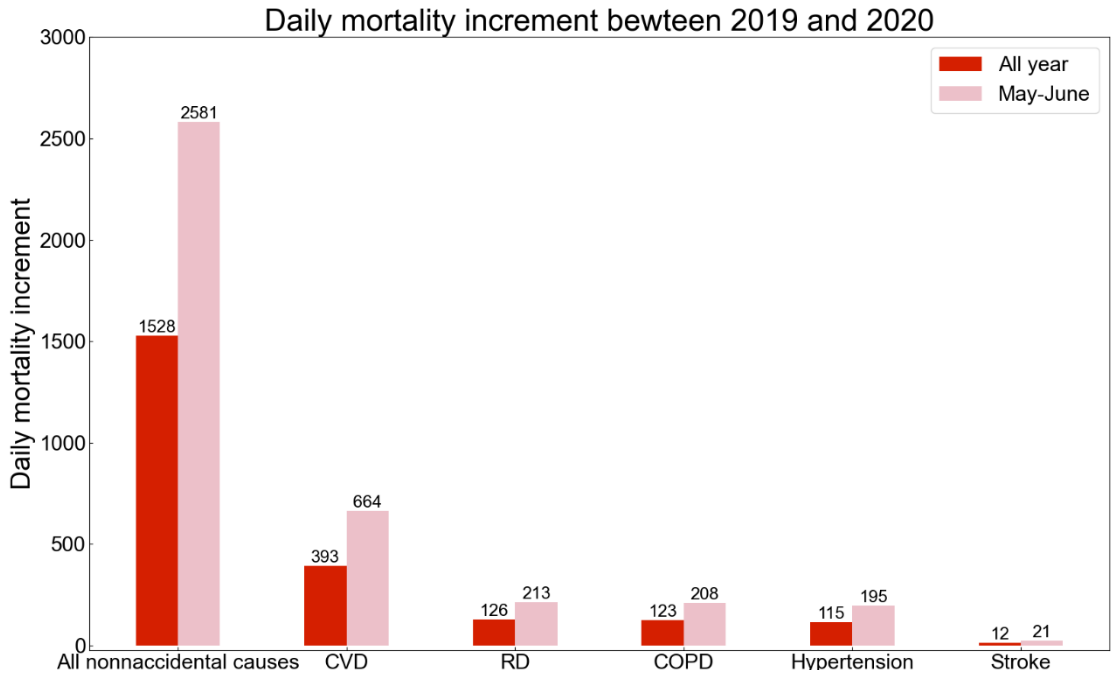

Figure S11. Difference in total daily mortality from all non-accidental causes, CVD, RD, COPD, hypertension, and stroke attributable to ambient O_3 exposure in all cities over the SCB within May-June and the whole year between 2019 and 2020.

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